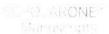
Environmental Science & Technology Letters

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Legacy and emerging perfluoroalkyl substances are important drinking water contaminants in the Cape Fear River Watershed of North Carolina

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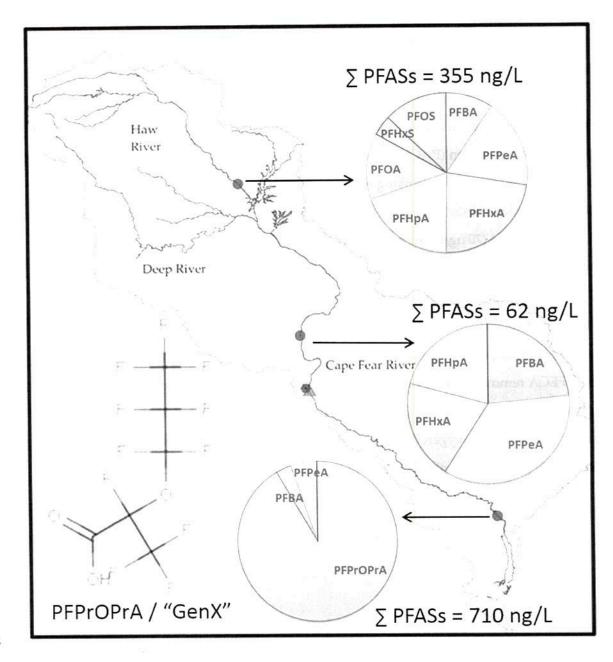


1	Legacy and emerging perfluoroalkyl substances are important
2	drinking water contaminants in the Cape Fear River Watershed of
3	North Carolina
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30	Long-chain perfluoroalkyl substances (PFASs) are being replaced by short-chain PFASs and
31	fluorinated alternatives. For ten traditionally studied PFASs and seven recently discovered
32	perfluoroalkyl ether carboxylic acids (PFECAs), we report (1) occurrence in the Cape Fear River
33	(CFR) watershed, (2) fate in drinking water treatment processes, and (3) adsorbability on
34	powdered activated carbon (PAC). In the headwater region of the CFR basin, PFECAs were not
35	detected in the raw water of a drinking water treatment plant (DWTP), but concentrations of
36	traditionally studied PFASs were high. The US Environmental Protection Agency's lifetime
37	health advisory level (70 ng/L) for perfluorooctane sulfonic acid and perfluorooctanoic acid
38	(PFOA) was exceeded on 57 of 127 sampling days. In raw water of a DWTP downstream of a
39	PFAS manufacturer, the mean concentration of perfluoro-2-propoxypropanoic acid
10	(PFPrOPrA), a replacement for PFOA, was 631 ng/L (n=37). Six other PFECAs were detected
11	with three exhibiting chromatographic peak areas up to 15 times that of PFPrOPrA. At this
12	DWTP, PFECA removal by coagulation, ozonation, biofiltration, and disinfection was
13	negligible. PFAS adsorbability on PAC increased with increasing chain length. Replacing one
14	CF2 group with an ether oxygen decreased PFAS affinity for PAC, while replacement of
15	additional CF2 groups with ether oxygens did not lead to further affinity changes.

47 Table of Contents Graphic



Introduction

30	Perfluoroalkyl substances (PFASs) are extensively used in the production of plastics, water/stail
51	repellents, firefighting foams and food-contact paper coatings. The widespread occurrence of
52	PFASs in drinking water sources is closely related to the presence of industrial sites, military
53	fire training areas, civilian airports, and wastewater treatment plants.1 Until 2000, long-chain
54	PFASs, such as perfluorocarboxylic acids (PFCAs) with 7 or more carbon atoms and
55	perfluorosulfonic acids (PFSAs) with 6 or more carbon atoms, were predominantly used.2
56	Accumulating evidence about ecotoxicological and human health effects ^{3, 4} associated with
57	exposure to long-chain PFASs has led to increased regulatory attention. Recently the U.S.
58	Environmental Protection Agency (USEPA) established a lifetime health advisory level (HAL)
59	of 70 ng/L for the sum of perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid
60	(PFOS) concentrations in drinking water. 5, 6 Over the last decade, production of long-chain
61	PFASs has declined in Europe and North America, and manufactures are moving towards
62	short-chain PFASs and fluorinated alternatives.7-10 Some fluorinated alternatives were recently
63	identified,8,11 but the majority of the organofluorine loading to the aquatic environment remains
54	unidentified. ¹²⁻¹⁴
65	One group of fluorinated alternatives, perfluoroalkyl ether carboxylic acids (PFECAs), was
66	recently discovered in the Cape Fear River (CFR) downstream of a PFAS manufacturing
57	facility. ¹¹ Identified PFECAs included perfluoro-2-methoxyacetic acid (PFMOAA), perfluoro-3-
68	methoxypropanoic acid (PFMOPrA), perfluoro-4-methoxybutanoic acid (PFMOBA), perfluoro-
59	2-propoxypropanoic acid (PFPrOPrA), perfluoro(3,5-dioxahexanoic) acid (PFO2HxA),
70	perfluoro(3,5,7-trioxaoctanoic) acid (PFO3OA) and perfluoro(3,5,7,9-tetraoxadecanoic) acid
71	(PFO4DA) (Table S1 and Figure S1 in supporting information (SI)). The ammonium salt of
'2	PFPrOPrA is a known PFOA alternative that has been produced since 2010 with the trade name
'3	"GenX". 15 According to the manufacturer, the ether oxygen enables "very rapid
4	bioelimination" ¹⁵ ; however, except for the PFPrOPrA data reported by the manufacturer, ¹⁶⁻¹⁸
5	little information is available on pharmacokinetic behavior, toxicity, or environmental fate and
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76	transport of PFECAs. To the knowledge of the authors, the only other published PFECA
77	occurrence data are for PFPrOPrA in Europe and China,19 and no published data are available

- 78 on the fate of PFECAs during water treatment.
- 79 The strong C-F bond makes PFASs refractory to abiotic and biotic degradation,²⁰ and most
- 80 water treatment processes are ineffective for legacy PFAS removal.²¹⁻²⁶ Processes capable of
- 81 removing PFCAs and PFSAs include nanofiltration,²⁷ reverse osmosis²⁴, ion exchange,^{27, 28} and
- 82 activated carbon adsorption,^{27, 28} with activated carbon adsorption being the most widely
- 83 employed treatment option.
- 84 The objectives of this research were to (1) identify and quantify the presence of legacy PFASs
- 85 and emerging PFECAs in drinking water sources, (2) assess PFAS removal by conventional and
- 86 advanced processes in a full-scale drinking water treatment plant (DWTP), and (3) evaluate
- 87 PFAS adsorbability by powdered activated carbon (PAC).

88 Materials and Methods

- 89 Water samples: Source water of three DWTPs treating surface water in the CFR watershed was
- 90 sampled between June 14 and December 2, 2013 (Figure S2 in SI). Samples were collected from
- 91 the raw water tap at each DWTP daily as either 8-hour composite (DWTP A, 127 samples) or 24-
- 92 hour composite (DWTP B, 73 samples; DWTP C, 34 samples). Samples were collected in 250-mL
- 93 HDPE bottles and picked up (DWTPs A and B) or shipped overnight (DWTP C) on a weekly
- 94 basis. All samples were stored at room temperature until analysis (within 1 week of receiving).
- 95 On August 18, 2014, grab samples were collected at DWTP C after each unit process in the
- 96 treatment train (raw water ozonation, coagulation/flocculation/sedimentation, settled water
- 97 ozonation, biological activated carbon (BAC) filtration, disinfection by medium pressure UV
- 98 lamps and free chlorine). Operational conditions of DWTP C on the sampling day are listed in
- 99 Table S2 in SI. Samples were collected in 1-L HDPE bottles and stored at room temperature
- until analysis. On the same day, grab samples of CFR water were collected in six 20-L HDPE

101 carboys at William O. Huske Lock and Dam downstream of a PFAS manufacturing site and 102 stored at 4°C until use in PAC adsorption experiments. Adsorption experiments: PFAS adsorption by PAC was studied in batch reactors (amber glass 103 104 bottles, 0.45 L CFR water). PFECA adsorption was studied at ambient concentrations (~1,000 105 ng/L PFPrOPrA, chromatographic peak areas of other PFECAs ~10-800% of the PFPrOPrA area). Legacy PFASs were present at low concentrations (<40 ng/L) and spiked into CFR water 106 107 at ~1000 ng/L each. Background water matrix characteristics are summarized in Table S3 in SI. A thermally-activated, wood-based PAC (PicaHydro MP23, PICA USA, Columbus OH, mean 108 diameter: 12 μ m, BET surface area: 1460 m²/g)²9 proved effective for PFAS removal in a prior 109 study²⁸ was used at doses of 30, 60 and 100 mg/L. These doses represent the upper feasible end 110 111 for drinking water treatment. Samples were taken prior to and periodically after PAC addition 112 for PFAS analysis. PFAS analysis: Information about analytical standards and the liquid chromatography-tandem 113 114 mass spectrometry (LC-MS/MS) method for PFAS quantification is provided in SI. 115 Results and Discussion 116 PFAS occurrence in drinking water sources: Mean PFAS concentrations in source water of 117 three DWTPs treating surface water from the CFR watershed are shown in Figure 1. In 118 communities A and B, only legacy PFASs were detected (mean ∑PFAS: 355 ng/L in community 119 A, 62 ng/L in community B). Detailed concentration data are shown in Table S6 and Figure S3 in 120 SI. In community A, PFCAs with 4-8 carbons, perfluorohexane sulfonic acid (PFHxS) and PFOS 121 were detected at median concentrations > QLs. Mean and median concentrations were 44 and 29 122 ng/L, respectively, for PFOS, and 46 and 34 ng/L, respectively, for PFOA. During the 127-day 123 sampling campaign, the sum concentration of PFOA and PFOS exceeded the USEPA HAL of 70 124 ng/L on 57 days, and the mean over the entire study period was 90 ng/L. Similar legacy PFAS 125 concentrations were observed in the same area ten years ago,30 suggesting that PFAS source(s)

upstream of community A have long-term negative impacts on drinking water quality. Also,

our data show that legacy PFASs remain surface water contaminants of concern even though their production has been phased out in the US. Relating total PFAS concentration to average daily stream flow (Figure S4 in SI) illustrated a general trend of low PFAS concentrations at high flow and high concentrations at low flow, consistent with the hypothesis of upstream point source(s). In community B, perfluorobutanoic acid (PFBA) and perfluoropentanoic acid (PFPeA) were the most frequently detected, with mean concentrations of 12 and 19 ng/L, respectively. Mean and median PFOA and PFOS concentrations were <QL, and the maximum sum concentration of PFOA and PFOS was 59 ng/L. Lower PFAS concentrations in community B relative to community A can be explained by the absence of substantive PFAS sources between the two communities, dilution by tributaries, and the buffering effect of Jordan Lake, a large reservoir located between communities A and B.

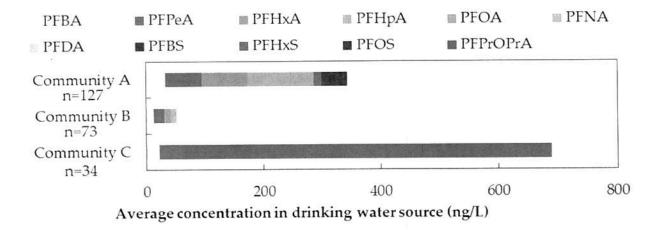
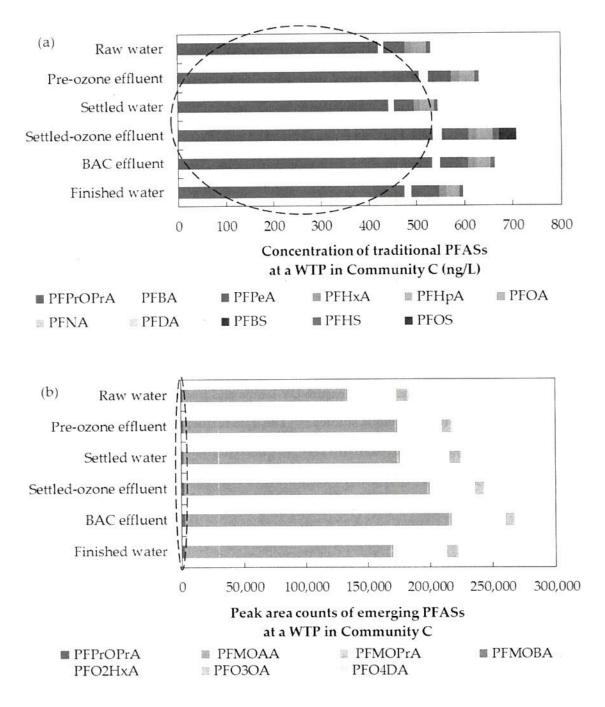


Figure 1. PFAS occurrence at drinking water intakes in the CFR watershed. Concentrations represent averages of samples collected between June and December 2013. Individual samples with concentrations < QLs were considered as 0 when calculating averages, and average concentrations < QLs were not plotted.

145	In community C (downstream of a PFAS manufacturing site), legacy PFAS concentrations were
146	low, and only mean (and median) concentrations of PFBA and PFPeA were >QLs. However,
147	high concentrations of PFPrOPrA were detected (up to ~4500 ng/L). The average PFPrOPrA
148	concentration (631 ng/L) was approximately eight times the average summed PFCA and PFSA
149	concentrations (79 ng/L). Other PFECAs had not yet been identified at the time of analysis.
150	Similar to communities A and B, the highest PFAS concentrations for community C were also
151	observed at low flow (Figure S3 in SI).
152	
153	PFAS fate in conventional and advanced water treatment processes: To investigate whether
154	PFASs can be removed from impacted source water, samples from DWTP C were collected at
155	the intake and after each treatment step. Results in Figure 2 suggest conventional and advanced
156	treatment processes (coagulation/flocculation/sedimentation, raw and settled water ozonation,
157	BAC filtration, disinfection by medium pressure UV lamps and free chlorine) did not remove
158	legacy PFASs, consistent with previous studies. ²¹⁻²⁵ The data further illustrate that no
159	measurable PFECA removal occurred in this DWTP. Concentrations of some PFCAs, PFSAs,
160	PFMOPrA, PFPrOPrA and PFMOAA may have increased after ozonation, possibly due to the
161	oxidation of precursor compounds.24 Disinfection with medium pressure UV lamps and free
162	chlorine (located the between BAC effluent and the finished water) may have decreased
163	concentrations of PFMOAA, PFMOPrA, PFMOBA and PFPrOPrA, but only to a limited extent.
164	Results in Figure 2 further illustrate that the PFAS signature of the August 2014 samples was
165	similar to the mean PFAS signature observed during the 2013 sampling campaigns shown in
166	Figure 1; i.e., PFPrOPrA concentrations (400-500 $\mu g/L$) greatly exceeded legacy PFAS
167	concentrations. Moreover, three PFECAs (PFMOAA, PFO2HxA and PFO3OA) ¹¹ had peak areas
168	2-113 times greater than that of PFPrOPrA (Figure 2b). The existence of high levels of emerging
169	PFASs suggests the necessity of incorporating them into routine monitoring.
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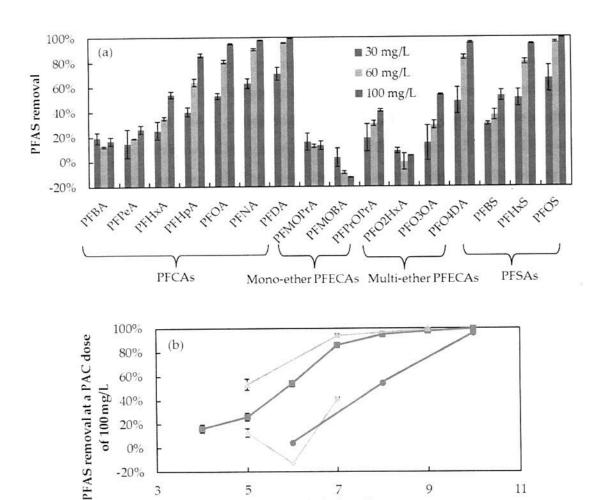
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Figure 2. Fate of (a) legacy PFASs and PFPrOPrA and (b) PFECAs through a full-scale water treatment plant. Because authentic standards were not available for emerging PFECAs, chromatographic peak area counts are shown in panel b. PFPrOPrA data are shown in both panels and highlighted in dashed ovals for reference. Compounds with concentrations <QL were not plotted.

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178	PEAS adsorption by BAC, BAC,
	PFAS adsorption by PAC: PAC can effectively remove long-chain PFCAs and PFSAs, but its
179	effectiveness decreases with decreasing PFAS chain length. ^{23, 24, 28} It is unclear, however, how the
180	presence of ether group(s) in PFECAs impacts adsorbability. After a contact time of 1 hour, a
181	PAC dose of 100 mg/L achieved >80% removal of legacy PFCAs with carbon chain length ≥7. At
182	a PAC dose of 60 mg/L, >80% removal was achieved for PFCAs with carbon chain length ≥8
183	over the same time. At a PAC dose of 100 mg/L, removals were 95% for PFO4DA and 54% for
184	PFO3OA, but <40% for other PFECAs. Detailed removal percentage data as a function of PAC
185	contact time are shown in Figure S5 in SI. PFMOAA could not be quantified by the analytical
186	method used in this test; however, based on the observations that PFAS adsorption decreases
187	with decreasing carbon chain length and that PFECAs with one or two more carbon atoms than
188	PFMOAA (i.e., PFMOPrA and PFMOBA) were poorly adsorbed by PAC (Figure 3), it is
189	expected that PFMOAA adsorption is negligible at the tested conditions.
190	To compare the affinity of different PFASs for PAC, the PFAS removal percentages in solution
191	were plotted as a function of PFAS chain length (the sum of carbon (including branched), ether
192	oxygen, and sulfur atoms) (Figure 3(b)). The adsorbability of both legacy and emerging PFASs
193	increased with increasing chain length. PFSAs were more readily removed than PFCAs of
194	matching chain length, which agrees with previous studies. ^{23, 24, 28} PFECAs exhibited lower
195	adsorbabilities than PFCAs of the same chain length (e.g. PFMOBA <pfhxa), suggesting="" td="" that<=""></pfhxa),>
196	the replacement of a CF2 group with an ether oxygen atom decreases the affinity of PFASs for
197	PAC. However, the replacement of additional CF2 groups with ether groups resulted in small or
198	negligible affinity changes among the studied PFECAs (e.g., PFMOBA~PFO2HxA).
199	Alternatively, if only the number of perfluorinated carbons were considered as a basis of
200	comparing adsorbability, the interpretation would be different. In that case, with the same
201	number of perfluorinated carbons, PFCAs have a higher affinity for PAC than mono-ether
202	PFECAs (e.g., PFPeA>PFMOBA), but a lower affinity than multi-ether PFECAs (e.g.,
203	PFPeA <pfo3oa).< td=""></pfo3oa).<>



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-20%

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→ PFCAs → Mono-ether PFECAs

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Figure 3. PFAS adsorption on PAC (a) at carbon doses of 30, 60 and 100 mg/L and (b) as a function of PFAS chain length. PAC contact time in CFR water was 1 hour. Legacy PFASs were spiked at ~1000 ng/L and the emerging PFASs were at ambient concentrations. Figures show average PFAS removal percentages, and error bars show one standard deviation of replicate experiments.

Chain length

--- Multi-ether PFECAs

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In either framework, it is clear that the presence of ether groups in PFECAs changes their propensity to leave the aqueous phase and adsorb on PAC. It can thus be inferred that the incorporation of ether groups changes physiochemical properties, such as the octanol-water

215	partition coefficient and aqueous solubility of PFECAs relative to PFCAs. Consequently, it is
216	reasonable to expect that fate and transport of PFECAs in natural and engineered systems will
217	differ from that of legacy PFCAs. For example, while PFPrOPrA ("GenX") may be less
218	bioaccumulative than PFOA, which it is replacing, the adsorption data here suggest PFPrOPrA
219	is less hydrophobic than PFOA. Thus, when released to the environment, PFPrOPrA has a
220	higher tendency to remain in the aqueous phase and is more difficult to remove from drinking
221	water sources by adsorptive means.
222	To the knowledge of the authors, this is the first paper reporting the behavior of recently
223	identified PFECAs in water treatment processes. We further show that legacy PFASs continue to
224	be a concern in the upper reaches of the CFR basin, and that PFECAs are an important class of
225	fluorinated alternatives that dominated the PFAS signature downstream of a fluorochemical
226	manufacturer. The relatively low concentrations of legacy PFASs in the finished drinking water
227	of community C are consistent with data reported from this DWTP in the third unregulated
228	contaminant monitoring rule (UCMR3) conducted by USEPA31. However, the detection of
229	potentially high levels of PFECAs, and the difficulty to effectively remove not only legacy
230	PFASs but also PFECAs with many water treatment processes, suggest the need for broader
231	discharge control and contaminant monitoring.
232	Acknowledgement
233	This research was supported by the National Science Foundation (Award # 1550222), the Water
234	Research Foundation (Project 4344), and the North Carolina Urban Water Consortium. The
235	views expressed in this article are those of the authors and do not necessarily represent the
236	views or policies of the USEPA.
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Legacy and emerging perfluoroalkyl substances are important drinking water contaminants in the Cape Fear River Watershed of North Carolina

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Supporting information includes analytical method description, 6 tables, and 5 figures.

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Analytical standards: PFASs studied in this research are listed in Table S1. For legacy PFASs, native and isotopically labeled standards were purchased from Wellington Laboratories (Guelph, Ontario, Canada). Native PFPrOPrA was purchased from Thermo Fisher Scientific (Waltham, MA). No analytical standards were available for other PFECAs.

PFAS quantification: PFAS concentrations in samples from DWTPs and adsorption tests were determined by liquid chromatography tandem mass spectrometry (LC-MS/MS) using a large-volume (0.9 mL) direct injection method. An Agilent 1100 Series LC pump and PE Sciex API 3000 LC-MS/MS system equipped with a 4.6 mm x 50 mm HPLC column (Kinetex C18 5μm 100Å, Phenomenex Inc.) was used for PFAS analysis. The eluent gradient is shown in Table S4 in SI. All samples, calibration standards, and quality control samples were spiked with isotopically labeled internal standards, filtered through 0.45-μm glass microfiber syringe filters, and analyzed in duplicate. The MS transitions for PFAS analytes and internal standards are shown in Table S5 in SI. The quantitation limit (QL) was 25 ng/L for PFOS and perfluorodecanoic acid, and 10 ng/L for other legacy PFASs and PFPrOPrA. For PFECAs without analytical standards, chromatographic peak areas are reported.

PFAS concentrations along the treatment train of DWTP C were analyzed using a Waters Acquity ultra performance liquid chromatograph interfaced with a Waters Quattro Premier XE triple quadrupole mass spectrometer (Waters, Milford, MA, USA) after solid phase extraction. Method details are described elsewhere. The QL for all PFASs with analytical standards was 0.2 ng/L, and peak areas were recorded for PFECAs without standards.

Table S1. Perfluoroalkyl substances (PFASs) detected in the Cape Fear River (CFR) watershed

Compound	Molecular weight	Formula	CAS#	# of perfluorinated carbons	Chain length (including all C, O and S)
Perflu	orocarboxylic	acids (PFCAs	s)		
Perfluorobutanoic acid (PFBA)	214.0	C4HF7O2	375-22-4	3	4
Perfluoropentanoic acid (PFPeA)	264.0	C5HF9O2	2706-90-3	4	5
Perfluorohexanoic acid (PFHxA)	314.1	C ₆ HF ₁₁ O ₂	307-24-4	5	6
Perfluoroheptanoic acid (PFHpA)	364.1	C7HF13O2	375-85-9	6	7
Perfluorooctanoic acid (PFOA)	414.1	C8HF15O2	335-67-1	7	8
Perfluorononanoic acid (PFNA)	464.1	C9HF17O2	375-95-1	8	9
Perfluorodecanoic acid (PFDA)	514.1	C10HF19O2	335-76-2	9	10
Perfl	uorosulfonic a	cids (PFSAs)			
Perfluorobutane sulfonic acid (PFBS)	300.1	C4HF9SO3	29420-49-3	4	5
Perfluorohexane sulfonic acid (PFHxS)	438.2	C6HF13SO3	355-46-4	6	7
Perfluorooctane sulfonic acid (PFOS)	500.1	C8HF17SO3	111873-33-7	8	9
Perfluoroalkyl ether carboxyl	ic acids with o	ne ether grou	ıp (mono-ether	PFECAs)	
Perfluoro-2-methoxyacetic acid (PFMOAA)	180.0	C ₃ HF ₅ O ₃	674-13-5	2	4
Perfluoro-3-methoxypropanoic acid (PFMOPrA)	230.0	C ₄ HF ₇ O ₃	377-73-1	3	5
Perfluoro-4-methoxybutanoic acid (PFMOBA)	280.0	C5HF9O3	863090-89-5	4	6
Perfluoro-2-propoxypropanoic acid (PFPrOPrA)	330.1	C6HF11O3	13252-13-6	5	7
Perfluoroalkyl ether carboxylic	acids with mu	tiple ether g	roup (multi-eth	er PFECAs)	
Perfluoro(3,5-dioxahexanoic) acid (PFO2HxA)	246.0	C ₄ HF ₇ O ₄	39492-88-1	3	6
Perfluoro(3,5,7-trioxaoctanoic) acid (PFO3OA)	312.0	C5HF9O5	39492-89-2	4	8
Perfluoro(3,5,7,9-tetraoxadecanoic) acid (PFO4DA)	378.1	C6HF11O6	39492-90-5	5	10

Table S2. Operational conditions of DWTP C on sampling day (August 18, 2014)

Parameter	Value
Raw water ozone dose	3.1 mg/L
Raw water total organic carbon concentration	6.0 mg/L
Aluminum sulfate coagulant dose	43 mg/L
Coagulation pH	5.70
Settled water ozone dose	1.3 mg/L
Settled water total organic carbon concentration	1.90 mg/L
Empty bed contact time in biological activated carbon filters	9.4 minutes for granular activated carbon layer 2.3 minutes for sand layer
Medium pressure UV dose	25 mJ/cm ²
Free chlorine dose	1.26 mg/L as Cl ₂
Free chlorine contact time	17.2 hours

Table S3. Water quality characteristics of surface water used in adsorption tests

Non-purgeable organic carbon (mg/L)	Ultraviolet absorbance at a wavelength of 254 nm	рН	Alkalinity (mg/L as CaCO ₃)	Conductivity (µS/cm)
9.036	0.399	7.53	19	133.5

Table S4. LC gradient method for PFAS analysis

Time (min)	Mobile Phase A% (v/v)	Mobile Phase B%	Flow Rate (mL/min)
0 – 2	95	5	0.9
2 – 5	95	5	0.9
5 – 10	95 → 10	$5 \rightarrow 90$	0.9
10 - 10.1	10	90	0.9
10.1 – 14	10 → 95	90 → 5	0.9

Mobile phase A: 2 mM ammonium acetate in ultrapure water with 5% methanol

Mobile phase B: 2 mM ammonium acetate in acetonitrile with 5% ultrapure water

Table S5. MS transitions for PFAS Analysis

Barrier principal de la constante de la consta	Compound	MS/MS Transition	Internal standard
	PFBA	212.8 → 168.8	13C4-PFBA
	PFPeA	262.9 → 218.8	13C2- PFHxA
	PFHxA	$313.6 \rightarrow 268.8$	13C2- PFHxA
	PFHpA	362.9 → 318.8	13C4- PFOA
Legacy PFASs	PFOA	413.0 → 368.8	13C4- PFOA
Legacy I FA3s	PFNA	463.0 → 418.8	13C4- PFOA
	PFDA	513.1 → 68.8	13C2-PFDA
	PFBS	299.1 → 98.8	18O2-PFHxS
	PFHxS	399.1 → 98.8	18O2-PFHxS
	PFOS	498.9 → 98.8	13C4-PFOS
PFECAs	PFMOAA	$180.0 \to 85.0$	N/A
	PFMOPrA	229.1 → 184.9	N/A
	PFMOBA	279.0 → 234.8	N/A
	PFPrOPrA	329.0 → 284.7	13C2- PFHxA
	PFO2HxA	245.1 → 85.0	N/A
	PFO3OA	311. → 84.9	N/A
	PFO4DA	377.1 → 85.0	N/A
	Perfluoro-n-[1,2,3,4- ¹³ C ₄]butanoic acid (13C4-PFBA)	217.0 → 172	
	Perfluoro-n-[1,2-13C2]hexanoic acid (13C2-PFHxA)	315.1 → 269.8	
	Perfluoro-n-[1,2,3,4-13C2]octanoic acid (13C4-PFOA)	417.0 → 372.0	N. 1. 11. 11.
nternal standards	Perfluoro-n-[1,2- ¹³ C ₂]decanoic acid (13C2-PFDA)	515.1 → 469.8	Not applicable
	Sodium perfluoro-1- hexane[18O2]sulfonate (18O2-PFHxS)	403.1 → 83.8	
	Sodium perfluoro-1-[1,2,3,4-13C4]octane sulfonate (13C4-PFOS)	502.9 → 79.9	

Table S6. Maximum, minimum, mean and median concentrations (ng/L) of PFASs in CFR watershed surface water as drinking water sources. *

		Community A				Community B			Community C			
	max	min	median	mean	max	min	median	mean	max	min	median	mean
PFBA	99	<10	26	33	38	<10	12	12	104	<10	12	22
PFPeA	191	14	44	62	38	<10	19	19	116	<10	30	36
PFHxA	318	<10	48	78	42	<10	<10	11	24	<10	<10	<10
PFHpA	324	<10	39	67	85	<10	<10	11	24	<10	<10	<10
PFOA	137	<10	34	46	32	<10	<10	<10	17	<10	<10	<10
PFNA	38	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
PFDA	35	<25	<25	<25	<25	<25	<25	<25	<25	<25	<25	<25
PFBS	80	<10	<10	<10	11	<10	<10	<10	<10	<10	<10	<10
PFHxS	193	<10	10	14	14	<10	<10	<10	14	<10	<10	<10
PFOS	346	<25	29	44	43	<25	<25	<25	40	<25	<25	<25
PFPrOPrA	<10	<10	<10	<10	10	<10	<10	<10	4560	55	304	631
PFOA+PFOS	447	0	64	90	59	0	0	9	55	<10	<10	<10
Σ PFASs**	1502	18	212	355	189	0	47	62	4696	55	345	710

^{*} Concentrations < quantification limits were considered as zero to calculate means and \sum PFASs.

^{**} Other PFECAs were present in water samples from community C but could not be quantified and were therefore not included in Σ PFASs

Figure S1. Molecular structures of PEFCAs in this study

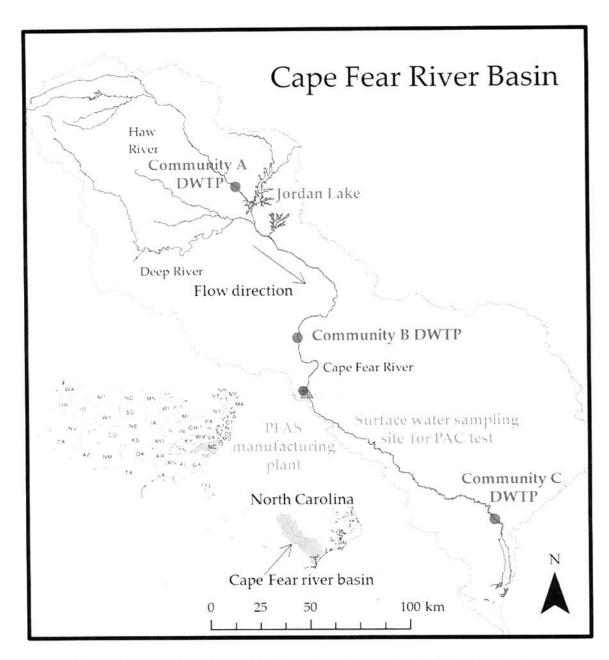
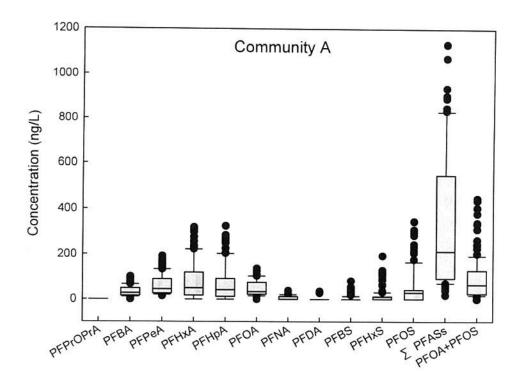
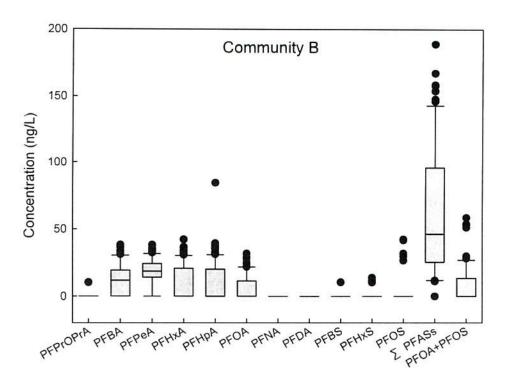


Figure S2. Sampling sites in the Cape Fear River watershed, North Carolina





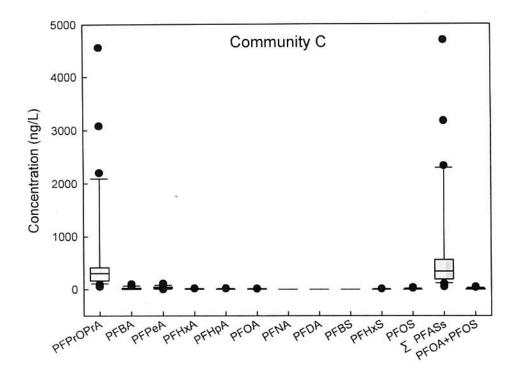


Figure S3. PFAS concentration distributions in the CFR watershed at three drinking water intakes. Concentrations < quantification limits were considered as zero. The upper and lower edges of a box represent the 75^{th} and 25^{th} percentile, respectively; the middle line represents the median; the upper and lower bars represent the 90^{th} and 10^{th} percentile, respectively; the dots represent outliners (> 90^{th} or < 10^{th} percentile).

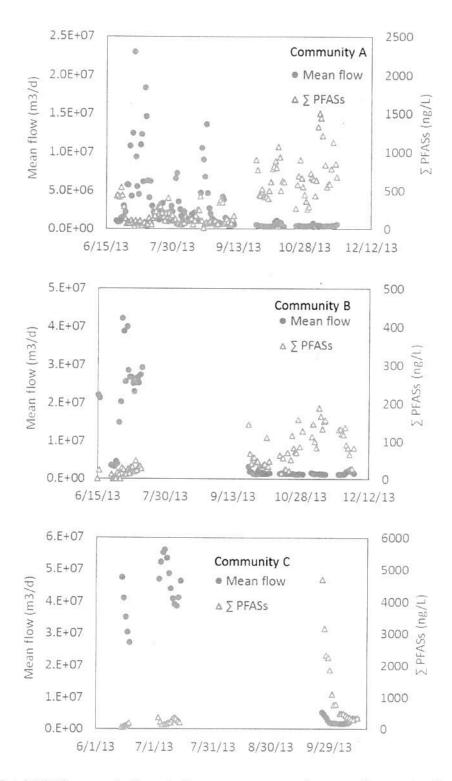


Figure S4. Total PFAS concentrations in the source water and stream flow at the three studied DWTPs. Stream flow data were acquired from US Geological Survey stream gage records

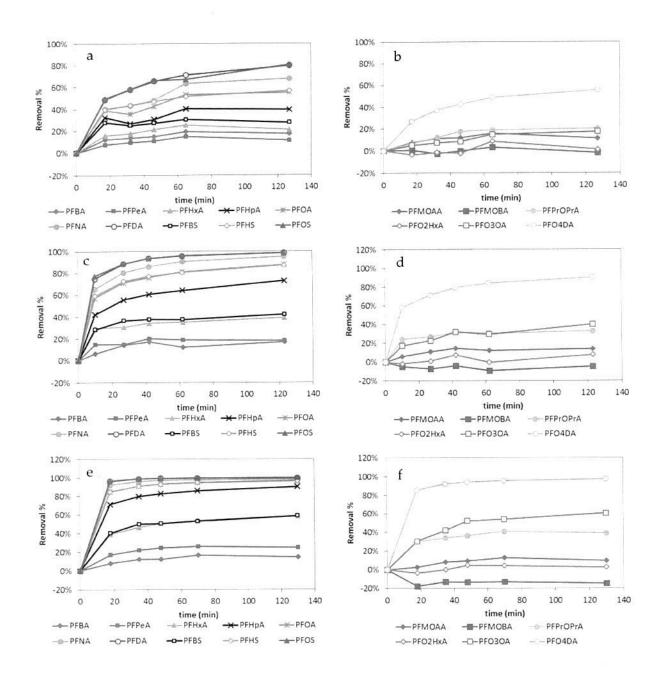


Figure S5. PFAS adsorption on PAC at carbon does of (a, b) 30 mg/L, (c, d) 60 mg/L and (e, f) 100 mg/L. Figures show average PFAS removal percentages of duplicate tests.

Reference

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